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Status of Source Apportionment Methods
Quail Roost II

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16. ABSTRACT

A source apportionment workshop sponsored by the United States Environmental Protection Agency was held at Quail Roost Conference Center in Rougemont, N.C. Thirty-five scientists from academic, governmental, and private institutions participated. The objective was to compare and document results of source apportionment analyses of simulated and real aerosol data sets. The simulated set was developed by scientists from the National Bureau of Standards and consisted of mass and elemental concentrations generated using a dispersion model that simulated transport of aerosols from a variety of sources to a receptor site. The real set contained the results of x-ray fluorescence, combustion, ion chromatography, x-ray diffraction, and scanning electron and optical microscopy analyses of aerosol samples collected in 18 12-hr sampling periods in Houston, TX. Ten groups analyzed these data sets using a variety of receptor models to apportion the simulated and real ambient measurements to potential contributing sources.

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STATUS OF SOURCE APPORTIONMENT METHODS: QUAIL ROOST II

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A source apportionment workshop sponsored by the United States Environmental Protection Agency was held at Quail Roost Conference Center in Rougemont, N.C. Thirty-five scientists from academic, governmental, and private institutions participated. The objective was to compare and document results of source apportionment analyses of simulated and real aerosol data sets. The simulated set was developed by scientists from the National Bureau of Standards and consisted of mass and elemental concentrations generated using a dispersion model that simulated transport of aerosols from a variety of sources to a receptor site. The real set contained the results of x-ray fluorescence, combustion, ion chromatography, x-ray diffraction, and scanning electron and optical microscopy analyses of aerosol samples collected in 18 12-hr sampling periods in Houston, TX. Ten groups analyzed these data sets using a variety of receptor models to apportion the simulated and real ambient measurements to potential contributing sources. The following recommendations and conclusions were derived from these workshop exercises: (1) Chemical mass balance and factor analysis methods could not easily resolve the data into more than eight categories; (2) chemical mass balance and ridge regression receptor models were able to resolve sources with similar elemental signatures; however, the effects of similarities in source signatures were manifest in the large uncertainties calculated for those source contributions; (3) source apportionment methods may need to be modified to incorporate chemical transformation of SO_2 and NO to sulfates and nitrates because current models tend to resolve particles into chemical species, rather than deducing their sources; and (4) scanning electron microscopy, optical microscopy, and X-ray diffraction methods are often needed to complement elemental-based receptor models in order to resolve sources having similar signatures.

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STATUS OF SOURCE APPORTIONMENT METHODS: QUAIL ROOST II

Introduction

On March 14-17, 1982, the "Mathematical and Empirical Receptor Models: Quail Roost II" Workshop was held in Rougemont, NC. Thirty-five scientists participated. The results of receptor modeling applied to real and simulated data sets were discussed, evaluated, and structured into components that would permit intercomparisons of the models. The participants also were asked to prepare a U.S. Environmental Protection Agency (EPA) report on the state of the art of receptor modeling.

The predecessor of this Quail Roost II Workshop was a March 1980 workshop also held at the Quail Roost Conference Center. At that 2½-day meeting (the "Quail Roost I Receptor Modeling Workshop"), scientists met to discuss the status of mathematical and analytical procedures to determine the natural and man-made sources that contribute to atmospheric particles. The objective was to obtain data and information for an EPA report that would describe the state of the art of receptor modeling (sometimes referred to as "source apportionment modeling"). This report was to contain recommendations for research projects that would advance knowledge in this relatively new area of air pollution science.

The EPA report that resulted from Quail Roost I[1] presented three major recommendations:

1. Develop a simulated air quality and source contribution data set perturbed by random error in order to test predictions of several different types of receptor models against known source contributions.

2. Obtain a real air quality data set to compare the consistency of receptor models and source apportionment procedures currently used by various groups in the United States.

3. Standardize validated models so that they can be readily implemented by Federal, state, and local agencies.

Following the Quail Roost I Workshop, several EPA and National Bureau of Standards (NBS) scientists worked to generate simulated and real air quality data sets consistent with Recommendations 1 and 2. The simulated set consisted of lists of mass and elemental concentrations for 40 sequential simulated fine particle aerosol observations at a single receptor site, and elemental composition profiles for 13 source types from which the observations were generated.

The real data set was derived from an EPA visibility study conducted in Houston, TX, during summer 1980. The data set was based on 18 sequential fine and coarse particle observations at a single field site, and included mass, elemental, and ionic species concentrations from beta gauge, x-ray fluorescence, neutron activation, and wet chemical analyses of the collection filters. In addition, filter samples from the Houston field study were subjected to x-ray diffraction (XRD), light microscopy (LM), and combined automated scanning electron microscopy (SEM)/x-ray fluorescence (XRF) analysis by selected participants.

Although the Houston aerosol was collected on three different filter media in order to permit a wide range of chemical analyses, the sampling protocol was not specifically designed for receptor modeling. For example, the filter mass loadings were a factor of ~2 less than adequate for XRD analysis. Davis of the South Dakota School of Mines and Technology (SDSM) (this volume) addresses this problem. Also, no special effort was made to collect and preserve the ambient nitrates. Ambient nitrates can be reliably measured with a denuder difference sampling system.[2] However, if particle sampling is conducted without these precautions, nitrate concentrations can be underestimated by a factor of 10. In spite of these sampling limitations, the Houston data set was considered to be an adequate and cost-effective basis for the Quail Roost II model intercomparison.

To complement the Houston ambient data, participants were provided with emissions inventories for Houston.[3] Unfortunately, emissions inventories are generally not adequate for receptor modeling as they seldom contain information on the elemental composition of the primary particle emissions or of the soil in the airshed under study. Source profiles specific to Houston, TX were not available. To redress this deficiency, source signatures from other cities[4,5] were provided. The participants were also permitted to use any other available source signatures, although this was not encouraged.

The availability of these data sets thus provided a basis for a second Quail Roost receptor model workshop. The principal objective of the Quail Roost II Workshop was to document the current status of source apportionment methodology by determining, for the simulated data set, how well the different receptor models were able to reproduce the true source contributions, and by comparing, for the real (Houston) data set, the consistency of results from different source apportionment methods when the true source contributions were unknown. A second objective was to determine the number of sources that could be reliably determined using only elemental compositional data and whether sources of similar emission characteristics could be resolved. A third objective was to delineate the weaknesses of receptor models and make recommendations to improve their performance.

The simulated and real data sets and associated source profiles were distributed in advance of the Quail Roost II Workshop on nine-track magnetic tape and in hard copy form. Between the time of receipt and the beginning of the Workshop (5 wk for the real data set, but less than 2 wk for the simulated set), the participating laboratory was expected to apply its particular source apportionment method to either or both data sets and prepare a report that would contain the following information:

1. Method(s) of source apportionment, including any special means for making judgments.

2. Estimates of the numbers and identities of source types.

3. For each source thought to be present, an estimate of its contribution ($\mu g/m^3$) to the individual periods.

4. Uncertainty estimates for each component, together with limits for components believed to be present but not detectable.

TABLE I. PARTICIPANTS WHO SUBMITTED RESULTS FOR THE SIMULATED AND REAL AIR QUALITY DATA SETS

Participant	Method	DATA SET	
		Simulated	Real
Environmental Research and Technology, Inc. (ERT)	Effective variance CMB	X	X
Nuclear/Environmental/Analysis, Inc. (NEA)	Effective variance CMB	X	X
U.S. Environmental Protection Agency (EPA) (Environmental Sciences Research Laboratory (ESRL)	Effective variance CMB/factor analysis	X	
Radian Corporation (RC)	Weighted least-squares CMB (real set) amd weighted ridge regression CMB (both sets)	X	X
Texas Air Control Board (TACB)	Multiple linear regression by tracers/factor analysis	X	X
South Dakota School of Mines and Technology (SDSM)	X-ray diffraction-effective variance CMB		X
State University of New York at Syracuse (SUNY)	Q-mode factor analysis[6] of SEM/XRF data		X
Boeing Aerospace Company, Seattle, WA (BC)	Light microscopy of coarse fraction		X
Lawrence Berkeley Laboratory (LBL)	X-ray diffraction		X
Harvard University (HU)	Weighted CMB/factor analysis	X	
University of Illinois (UI)	Target transformation factor analysis	X	X

Discussion of Results

The key Quail Roost II participants who submitted results for the simulated and real data sets are listed in Table I.

For both simulated and real data sets, the University of Illinois and Harvard University submitted factor analysis results, the Texas Air Control Board submitted multiple linear regression results, and Environmental Research and Technology, Inc., Radian Corporation, and Nuclear/Environmental/Analysis, Inc. submitted results based on chemical mass balance models. EPA submitted factor analysis results for one of the simulated data sets. Table I lists these and other contributions to the analysis.

Simulated Data Set

NBS sent to the participants three simulated data sets. However, only sets I and II were analyzed by the participants, since they had only 2 wk prior to the Workshop to prepare their reports.

Data set I was made up of 40 observations along with elemental source signatures for 13 potential contributing sources. (Only eight of these were in fact active.) Unknown to the participants, an additional unspecified source also contributed to the set I observations. Data set II also consisted of 40 observations. (All 13 known sources contributed.) Each data set contained at least four source groups within which signatures were similar. Sets I and II were designed to determine whether any of the receptor models could resolve sources emitting particles of similar elemental composition, and whether any model could detect and calculate the mass contributed to the receptor site by an unknown source.

None of the conventional chemical mass balance (CMB) methods detected the unknown source, but two of the factor analysis methods and a multilinear regression model were able to deduce the mass contribution of the unknown source. All of the receptor models had difficulty resolving sources with similar signatures. These difficulties created substantial uncertainties in the apportionment of those sources. In a companion paper (this volume), Currie of NBS presents additional details on the development of the simulated data and the participants' results for those data.

Real Data Set

Figure 1 shows that the conventional receptor or factor analysis models could only apportion the fine and coarse into three chemical categories (carbon, nitrate, and sulfate) and four general sources (vehicle exhaust, steel, marine, and crustal). On the other hand, LM and automatic SEM/XRF analysis were able to resolve 18 sources. Even though the sample mass loadings were inadequate for XRD, participants from Lawrence Berkeley Laboratory and SDSM were able to determine that most of the calcium in the coarse particles was of local soil origin based on their identification of a form of $CaCO_3$ indigenous to Houston.

Dzubay et al. (this volume) detail the results of the receptor model intercomparison based on the Houston data.

Considerations in Design of Source Apportionment Studies

A group of participants headed by Gordon of the University of Maryland examined and evaluated the preliminary results presented at the Workshop and prepared reports that contained (1) an assessment of the value of the real and simulated sets, and (2) a series of recommendations that serve as a basis for design of source apportionment studies by private or governmental institutions. Their findings and recommendations, prepared principally by Gordon with input from other Quail Roost II participants, appear in forthcoming report on the Workshop.[7] One of the key issues addressed by this Workshop subgroup was the development of protocols for data collection and analysis appropriate to different levels of source apportionment. The various levels of effort listed below assumes the source apportionment study is being implemented to address the issue of developing air pollution control strategies to reduce particulate mass loadings in an airshed to be in compliance with EPA's air quality standard for particulate matter. Therefore, the site where samples are to be collected and analyzed for receptor modeling are located where particulate mass has been observed to be in violation of the standard. The elements (and/or chemical species) that would be measured as input to the receptor model would vary from airshed to airshed but some species that are typically determined are listed in Table III. The Quail Roost II participants generally recommended that source apportionment studies be divided into three levels of intensity based principally on resource availability:

Level I: Minimal Resources. This effort uses existing source signature libraries, existing ambient air quality data (mass and elemental composition of fine and coarse particles), local emissions inventories, and literature values for soil composition. These data suffice for an initial apportionment by CMB and for factor analysis to determine if there are contributing sources that are not included in the emissions inventory. The cost for this level of effort is between $10-25,000, depending on whether or not additional sample analysis is required. Most of the effort will be spent gathering emission data and inputting data to the receptor model.

Level II: Moderate Resources. This level of effort includes all of the Level I work plus source elemental profiles from grab samples of fugitive sources, soil, and major primary particle emitters in the airshed. These grab samples are resuspended and recollected in the same manner and on the same filter material used to collect particles at the receptor site(s). Most of the expenditures for level II recptor modeling study is in the collection, redeposition, and analysis of grab samples. Level II costs could range from $25-100,000, depending on the size of the airshed under study.

Level III: Maximum Resources. This level of effort calls for (1) collection of aerosols at receptor sites where maximum plume fumigation may occur; (2) collection of particle emissions from the major emission sources with a dilution sampler that is coupled with a sampler similar to those used at the receptor sites; (3) analysis of filters from the sources and receptor sites by XRF and instrumental neutron activation analysis; (4) use of XRD, SEM, and LM to resolve sources with similar emission profiles. This level of effort could cost between $100-500,000, again depending on the size of the airshed under investigation. Most of the costs are in the collection and analyses of source and ambient samples.

TABLE II. SUGGESTED INSTRUMENTATION TO COLLECT AMBIENT DATA FOR RECEPTOR MODELING

Particulate Sampler	Flow Rate (liters/min)	Filter Medium	Species Measured	Method
Dichotomous	17	Teflon	Mass	β gauge
			Elements	XRF, INAA
		Quartz	Carbon	Combustion
			Ionic species (SO_4^{2-}, NO_3^-, etc.)	Ion chromatography/chemistry
		Nuclepore	Individual particles	SEM/XRF
Dichotomous	50	Teflon	Crystals, minerals	XRD, LM
Hi-Vol (with cyclone) 2.5 μm cutpoint	500-1000	Quartz	Organic compounds	gas chromatography/ mass spectrometry, Liquid Liquid Chromatography, etc.

Gas Sampler	Gases Measured	Output
Total sulfur	SO_2 and other sulfur-containing gases	Real time, need data-logging system
NO_x	NO, NO_2	Real time, need data-logging system
O_3	O_3	Real time, need data-logging system
Polyurethane	Organic vapors	Later extraction and analysis in laboratory

Other Instrument	Parameter Measured	Output
Meteorological tower	Wind speed, wind directions, temperature, dew point	Real-time, need data-logging system

TABLE III. SOURCE CATEGORIES AND MARKER ELEMENTS USED IN SOURCE APPORTIONMENT OF HOUSTON AEROSOL

Marker Elements	General Source Category	Specific Source Categories
Na, Cl	Marine	Marine
Al, Si, K, Ca, Mn, Fe	Crustal	Wind erosion of soil Paved and unpaved roads Construction Limestone crushing and handling Cement Lime kiln Fly ash Slag
C, Br, Pb	Transportation	Noncatalyst vehicles Catalyst cars Diesels
Mn, Fe	Steel	Iron and steel production Steel finishing and handling
Cu, Zn, As, Sm, Sb	Nonferrous metals	
NH_4^+, SO_4^{2-}	Sulfate	Primary emissions in Houston airshed Conversion of SO_2 in Houston airshed Regional background
C	Carbonaceous	Refineries Botanical Vegetative burning Tire wear
NO_3^-	Nitrate	Photochemical

The subgroup compiled a list (Table II) of aerosol sampling and gas measurement methods that should be part of any station that collects ambient data for receptor modeling purposes. Figure 2 depicts the aerosol sampling and analysis protocol that would be followed in a complete aerosol apportionment study. Note that at least four different sampling procedures may be necessary to perform reliable source apportionments. Gordon's subgroup further recommended that multivariate and wind trajectory receptor models be used whenever there is a need to obtain source signatures not readily available by conventional source sampling procedures. These models can provide source signatures by their unique treatment of air quality data. However, multivariate statistical models require large data sets (40 or more observations), and wind trajectory methods require short sampling periods (6 hr or less) and appropriate meteorological measurements.

Recommendations for Future Research

Table III lists the marker elements used by the Quail Roost II participants in their various receptor models to report the emission sources contributing to ambient particles impacting our Houston receptor site. The sulfate and nitrate listed in Table III do not, in the absence of appropriate source signatures, have marker elements that reliably indicate their origins. Even analysis of the Houston filters by SEM and LM failed to deduce the origins of the sulfate and nitrate. In most airsheds in the United States, sulfate and nitrate and their associated cations represent a substantial portion of the particle mass below 10 μm. Procedures must be developed to determine the origins of sulfate and nitrate that impact a given receptor site if source apportionment methodology is to adequately address urban and rural particulate problems.

At Quail Roost I and II, no participant presented methods to incorporate within the receptor modeling framework procedures to deal with nonconserved species, particularly sulfates and nitrates. In view of the dominance of these species in urban and rural aerosols, this issue requires resolution. Since Quail Roost II Lewis et al. (1983) have proposed a receptor model theory of secondary species which has the general form:

$$M_j = S_j \cdot R_j \cdot T \qquad (1)$$

where: M_j = mass concentration of secondary particles (e.g., $SO_4^=$) from source j impacting receptor site.

S_j = mass concentration of primary fine particles from source j impacting receptor site. This term is derived from CMB calculations.

R_j = ratio of SO_2 to fine particle emissions measured at source j.

T = transformation/deposition function [i.e., loss of SO_2 enroute from source j to receptor site, due to SO_2 dry deposition and $SO_4^=$ conversion]. T requires only average rate values for these processes.

A key assumption underlying equation (1) is that SO_2, primary fine particles, and secondary sulfate have identical behavior with regard to dispersion, but that SO_2 may experience additional dry deposition loss beyond that for the other two aerosol species. The application of equation (1) requires that back trajectory meteorological data are available.

Conclusions

The Quail Roost II Workshop exercises led to a number of conclusions that enhance our understanding of source apportionment methodologies. Some of the most important are as follows:

1. Multivariate procedures isolated at most seven of the nine sources active in simulated data set I. For simulated data set II, target transformation factor analysis isolated only four of the 13 sources.

2. The simulated data set exercises indicate that neither a multivariate nor CMB approach to source apportionment can be considered the method of choice for all circumstances. While a CMB approach is mathematically easier to implement and does not require a large number of samples, it lacks credibility in circumstances where the sources and their compositions are not well known. On the other hand, while multivariate analysis does not require comprehensive information on source signatures in advance, it does require a very large number of samples, and it appears that only four to six classes of sources can be resolved in a multisource airshed.

3. The unavailability of source profiles from the major emission sources in Houston and the absence of adequate soil samples limited the resolving power of the source apportionment models as applied to real data set.

4. LM and SEM successfully resolved the general categories of the Houston samples into several specific source related categories. The integration of these methods into a mathematical receptor model has not been demonstrated.

5. XRD appears to be a viable alternative to other analytical techniques for identifying certain crustal and combustion sources. However, this alternative was not adequately demonstrated with the Houston data set because special sampling techniques tailored to XRD were not used in collecting those samples.

6. Current models, which tend to resolve particles into chemical categories and not to deduce their sources, need to be modified to incorporate chemical transformation of SO_2 and NO into secondary products.

Recommendations

The major recommendations listed below are based on the Quail Roost II Workshop participants' evaluation of the receptor model intercomparisons and of their past experience with air quality source apportionment studies conducted for state and local agencies.

1. EPA should develop a well documented source apportionment package to assist Federal, state, and local agencies involved in air quality management. This package should include multiple mathematical options for performing source apportionments. The selection of a given option would depend on the resources and ambient and source data that are available. One option, for example, would be to perform factor analysis on receptor samples, followed by source sampling (as indicated by the factor analysis), followed in turn by CMB analysis. Selected ambient samples might then be analyzed by LM as a quality assurance check on the apportionment of the coarse particles.

2. The categories for reporting results from receptor modeling need to be standardized. The source profiles should be given (or referenced to the literature) along with the source intensities derived from them. One of the difficulties in intercomparing the Quail Roost II results was the ambiguity of some of the source categories used by different participants in their analyses of the Houston data set.

3. A source signature library needs to be established for receptor modeling studies. Each signature must be obtained in a receptor-compatible manner. That is, the source sample must be collected in such a way that the properties of the sample at the source and at the receptor are as similar as possible, and the analytical techniques used on the source and receptor samples must be essentially identical. These criteria may be met by dilution sampling of source emissions in which the aerosol is collected on filter media and with collection devices equivalent to those used at the receptor site.

4. Conduct a Quail Roost III Workshop in which the various source apportionment methods are applied to air quality and source signature data obtained by EPA in 1982 for the Philadelphia airshed. The Philadelphia source signature and air quality measurements were obtained by sampling and analysis procedures recommended in this report. Participants of Quail Roost III should be provided with the analytical data at least 9 mo prior to the workshop to allow sufficient time for analysis and report preparation.

5. The results of receptor and dispersion modeling should be assessed in concert to refine and improve the accuracy of both approaches.

Disclaimer

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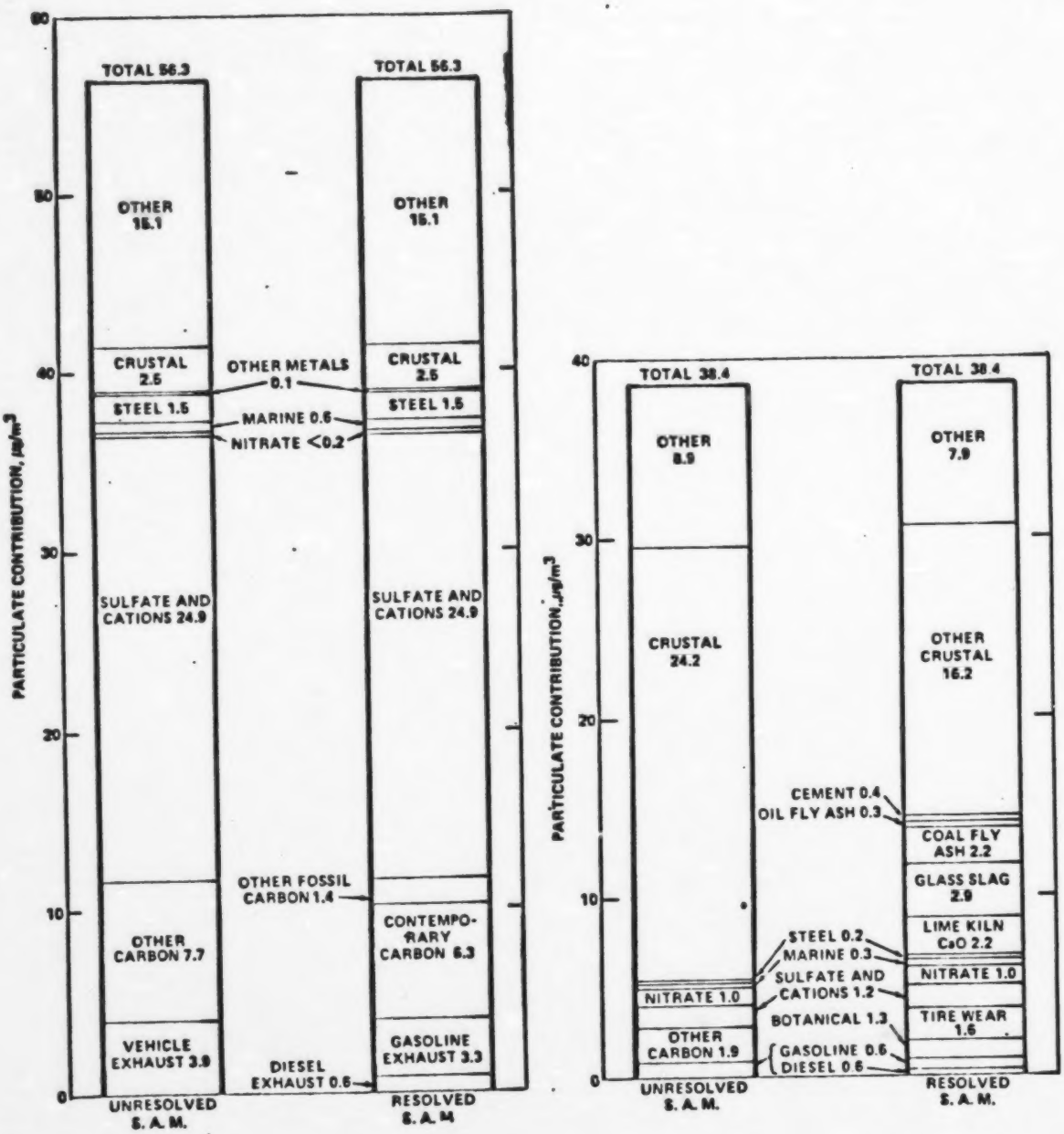

Figure 1. Results of source apportionment of real (Houston) data set. Left graph: fine fraction. Right graph: coarse fraction. S.A.M. = source apportionment method

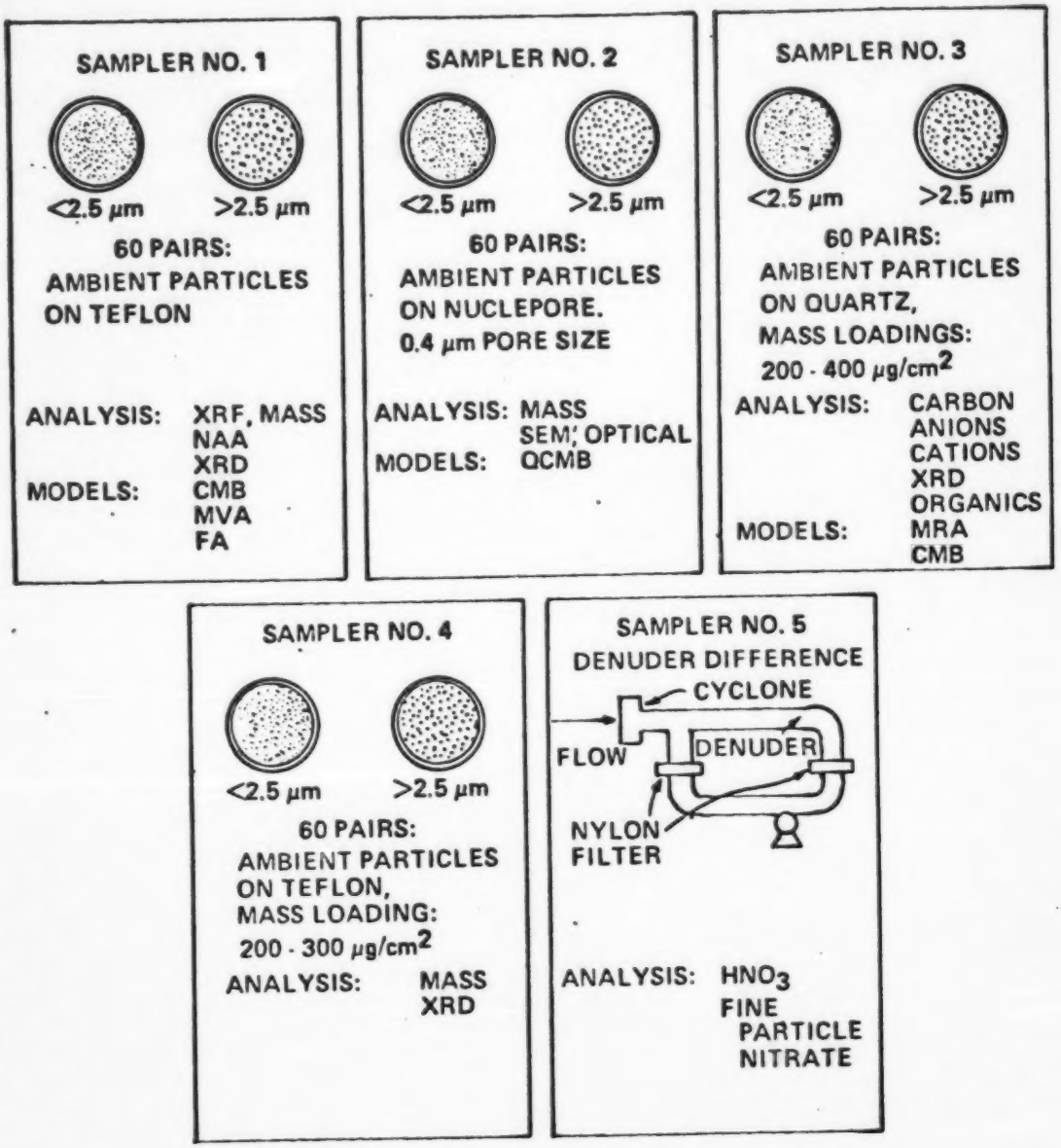

Figure 2. Ambient sampling and analysis protocol for a complete aerosol apportionment study.